

Nuclear forensic science—From cradle to maturity

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Abstract

Since the beginning of the 1990s, when the first seizures of nuclear material were reported, the IAEA recorded more than 800 cases of illicit trafficking of nuclear or other radioactive materials. Despite the decreasing frequency of seizures involving nuclear materials (i.e. uranium or plutonium), the issue continues to attract public attention and is a reason for concern due to the hazard associated with such materials. Once illicitly trafficked nuclear material has been intercepted, the questions of its intended use and origin are to be addressed. Especially the origin is of prime importance in order to close the gaps and improve the physical protection at the sites where the theft or diversion occurred. To answer the questions, a dedicated nuclear forensics methodology has been developed. In this paper, an overview is given on the methodologies used, the measurement techniques that are applied and on the characteristic parameters that help in the identification of the origin of the material. Some selected examples shall illustrate the challenges and the complexity associated with this work. In particular the past and on-going developments in this new area of science will be highlighted and special attention is attributed to the challenges ahead.

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1. Introduction

With the breakdown of the Soviet Union in the early 1990s a new phenomenon was observed, the so-called “nuclear smuggling”. The first cases ever were reported in 1991 in Switzerland and in Italy. In the subsequent years numerous incidents involving radioactive or nuclear material were reported from Germany, Czech Republic, Hungary and other central European countries. Apart from the need for determining the nature of the material, the authorities expressed interest in learning more about the intended use of the seized material, about its origin and about its potential trafficking route. In consequence, nuclear measurement laboratories and research institutes were confronted with the need for analysis of these materials and for data interpretation. A new branch of science was born: “nuclear forensics”. Analytical methods were mostly borrowed from the nuclear safeguards area and complemented with some material science investigations. Data interpretation was essentially based on the know-how and on the expertise of knowledgeable individuals and on information available in the open literature. In the mid 1990s the laboratories involved in these nuclear foren-

sic investigations started cooperating on an international level, particularly in the “Nuclear Smuggling International Technical Working Group” (ITWG). The analytical approach became more and more systematic and new methodologies were developed and implemented. Model calculations (e.g. burn-up calculations using codes like SCALE or ORIGEN) were used for determination of the mode of production of plutonium. A nuclear materials database was set up in a bilateral collaboration between the Bochvar Institute in Moscow and the Institute for Transuranium Elements (ITU) in Karlsruhe. This database serves for guiding the analysis and for attribution of materials.

As in classical forensics, also nuclear forensics relies on the fact that certain measurable parameters in a sample are characteristic for the given material. Using these characteristic parameters, one can draw conclusions on the intended use and on the possible origin of the material. The first step, however, consists in the identification of those parameters that are determined by the fabrication process or by the starting material, as only these parameters will provide useful information.

2. Characteristic parameters

This section focuses on the characteristic parameters or “signatures” found in the nuclear material, which may lead to reveal

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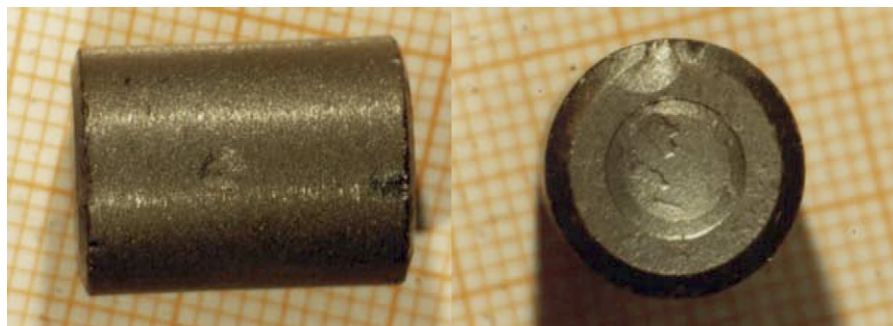


Fig. 1. Photo of a U pellet of “Find-1”.

the origin of the material. As these parameters have been identified and made use of subsequently, they will be described in the chronological sequence of their application to the area of nuclear forensics. A detailed description of the analytical techniques used to determine these signatures can be found elsewhere [1,2]. The signatures are presented together with examples of real seized nuclear materials, which demonstrate well the information obtained from different types of samples.

2.1. Major elemental and macroscopic composition

In March 1992 seized nuclear material was analysed at the ITU for the first time. It marked the birth of a new discipline in science: nuclear forensics. This first seizure, the so-called “Find-1” was intercepted in Augsburg, Germany and it consists of 72 uranium pellets (Fig. 1). The investigations were performed using standard techniques as applied for safeguards measurements: potentiometric titration for determining the uranium content, thermal ionisation mass spectrometry (TIMS) for measuring the isotopic composition and optical microscopy for determining the macroscopic parameters. Data interpretation and source attribution were essentially based on the expertise of senior scientists and of knowledgeable individuals. Comparative data had to be searched in the open literature.

The analysis of “Find-1” resulted in the following data (Table 1).

One can notice that the analytical effort at the time was fairly small. However, these results were sufficient to attribute the intended use of the material being fuel pellets for a Russian type graphite moderated reactor (RBMK). They also gave a clue about the possible origin of the material this being either UMP (Ulba Metallurgy Plant) in Kazakhstan or Elektrostal in Russia.

This is a good example illustrating how simple parameters like physical dimensions and isotopic composition of the major

elements of nuclear materials, i.e. U and Pu, reveals the intended use of the material. Nuclear materials can be divided into three categories depending on the abundance of their main fissionable isotope (i.e. ^{235}U or ^{239}Pu). These are: weapons grade material (uranium with an isotopic abundance of $^{235}\text{U} > 90\%$ or plutonium with an isotopic abundance of $^{240}\text{Pu} < 7\%$), weapons utilisable material ($^{235}\text{U} > 20\%$) and reactor grade material ($^{235}\text{U} < 20\%$, $^{240}\text{Pu} > 19\%$) [3].

In case of pellets the dimensions are an important additional parameter for determining the intended use. The dimensions are characteristics for different reactor types, varying significantly from reactor type to another. Thus they can clearly point to a certain reactor type. For unknown nuclear material mainly consisting of Pu, its isotopic composition may reveal the reactor type where the material was produced. Pu is produced as a by-product in a nuclear reactor; while neutron induced fission is the main reaction, neutron capture of uranium results in plutonium isotopes. Different reactor types use different initial ^{235}U enrichments as well as the neutron energy spectrum is different from reactor to reactor, and these two parameters cause significantly different Pu isotopic compositions [4–6]. Weapons grade Pu material typically is produced in reactors having very soft neutron spectra and in which the fuel can be continuously reloaded. These types of reactors are e.g. heavy-water and graphite moderated reactors. So-called reactor grade Pu ($^{240}\text{Pu} > 19\%$) is produced typically in light-water reactors after a few years irradiation [3]. This kind of Pu is not suitable for nuclear weapons. The content of U or Pu in a sample provides an indication about the stoichiometry of compound in question (e.g. uranium content in U_3O_8 is 84.8 wt.%, in UO_2 the uranium content is 88.15 wt.%) and about the chemical purity of the sample. Finding significantly lower concentration of the main element compared to the theoretical value points usually to large impurity content and, therefore

Table 1
Analytical results of “Find-1” (average of three pellets ± 1 s)

Parameter	Dimension	U-isotope (wt.%)	U-concentration (wt.%)
Ø (mm)	11.45 ± 0.01	U-232 ($8.27E-8 \pm 6.5E-9$)	87.98 ± 0.03
Height (mm)	14.42 ± 0.24	U-234 (0.034 ± 0.002)	
Weight (g)	15.454 ± 0.315	U-235 (2.507 ± 0.014)	
		U-236 (0.449 ± 0.051)	
		U-238 (97.011 ± 0.067)	

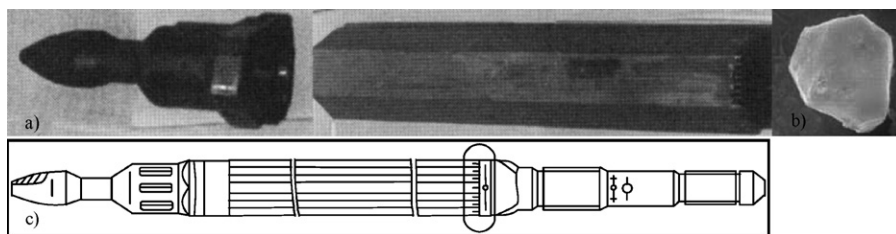


Fig. 2. (a) Contaminated metal pieces, (b) UO_2 particle ($\sim 8 \mu\text{m}$), and (c) drawing of the BN-600 fuel assembly.

indicating the material might be either an intermediate product or scrap.

Additives (e.g. Er, Gd and Ga) may be also found in U or Pu materials. They have usually concentrations in the low per cent range to, thus being distinctly different from “normal” impurities. Different additives serve for different purposes, e.g. erbium and gadolinium are used as burnable poisons in certain fuels [7,8], and gallium is used to stabilise the δ -phase of plutonium in large temperature range [9].

2.2. Minor elemental and microscopic composition

The continued interception of nuclear material and the need for more detailed information on the very nature of the material have triggered the further development of nuclear forensic science. The “adolescence” of this young discipline is illustrated in the following example. In February 1997 two pieces of radioactively contaminated metal were found at the scrap metal yard in Karlsruhe, Germany (Fig. 2a). On the swipe samples taken from the pieces, more than 10 UO_2 particles were found (Fig. 2b). The particle size varied between 2 and $10 \mu\text{m}$. The ^{235}U enrichment varied also between the particles, ranging from low enriched uranium (1.9 wt.%) to highly enriched uranium (89.1 wt.%). The variation in the ^{235}U enrichment can be explained by the possible contamination of the particles with natural uranium. Besides the uranium also traces of fission products were found, namely $^{134,137}\text{Cs}$. The analysis of the metal pieces showed that they were stainless steel. The elemental composition of the steel revealed its origin being either Eastern Europe or Russia. The dimensions of the pieces agreed with the upper and middle part of a drawing found in the open literature presenting the fuel assembly of BN-600 reactor, which is a Russian type fast breeder reactor (Fig. 2c).

The only BN-600 reactor, which is still operational and produces energy commercially, locates in Beloyarsk, Russia. This reactor, however, uses MOX (mixed U–Pu oxide) as a fuel and therefore, cannot be the origin of the fuel assembly. But similar assemblies are used also in the BR-10 research reactor in Russia, where the fuel, for testing purposes, can be enriched up to 90 wt.% in ^{235}U . This reactor located in Obninsk appears to be the last legal owner of the material.

This example illustrates the additional benefits obtained in nuclear forensic analysis with the introduction of microanalytical techniques, providing information on chemical and isotopic composition of individual particles, as well as on their morphology. Microanalytical techniques such as secondary ionisation mass spectrometry (SIMS) and scanning electron microscopy

(SEM) are powerful tools for particle and powder analysis. The elemental composition of particles can be determined by SEM coupled with electron-dispersive X-ray (EDX) system, and isotopic composition of U and Pu by SIMS or by the so-called fission-track method combined with TIMS [10–12]. Particularly, in case of powder samples, the question of material homogeneity arises, i.e. whether the sample consists of a single component or if it is a mixture of several components. In the latter case, the bulk analysis might result a wrong answer, if e.g. the isotopic composition of the different components is not the same.

SEM has also been applied for determining particle size distribution in powder samples and transmission electron microscopy (TEM) was used for grain size analysis.

2.3. Geolocation methods

After a decade of nuclear forensic investigations, a fair degree of maturity has been reached: analytical protocols are developed, handling procedures are in place and a data basis helping the interpretation has been established. In December 2003, nearly 3 kg of radioactive material was found in a shipment of scrap metal in the harbour of Rotterdam, The Netherlands (Fig. 3). The rather humid, yellowish coloured material was contained in a filter bag. In the same load also four process vessels were found, which were checked for contamination by taking a swipe sample. Two more swipes were taken from pipework, which was also part of the same shipload.

The material contained in the filter bag appeared to be natural uranium, which had obviously been processed, because no daughter products (e.g. radium and polonium) were found by gamma spectrometry. The U-content was fairly low, being only $35 \pm 2 \text{ wt.}\%$, which is very low value even for an intermediate product. However, when the material was dried in the oven, it lost 50% of its weight. Thus the U-content in dry material was around 70 wt.%, which is a typical value for a “yellow cake”. Due to the low U-content, the material contained lot of impurities. The following elements were found to be present as major impurities having concentrations $>1000 \mu\text{g/g}$ of material: Al, Ca, Cr, Fe, Mg, Mo, Na, Ni and P. Also the presence of some anions was checked qualitatively and NO_3^- and CO_3^{2-} were found. The “age” of the material was determined using the $^{234}\text{U}/^{230}\text{Th}$ parent daughter ratio. The apparent age was calculated to be 100 ± 10 years; this clear overestimation is due to incomplete separation of thorium during the last chemical purification.

The swipe samples from process vessels showed traces of natural uranium, whereas the swipes from process piping contained traces of fission products and transuranium elements, such

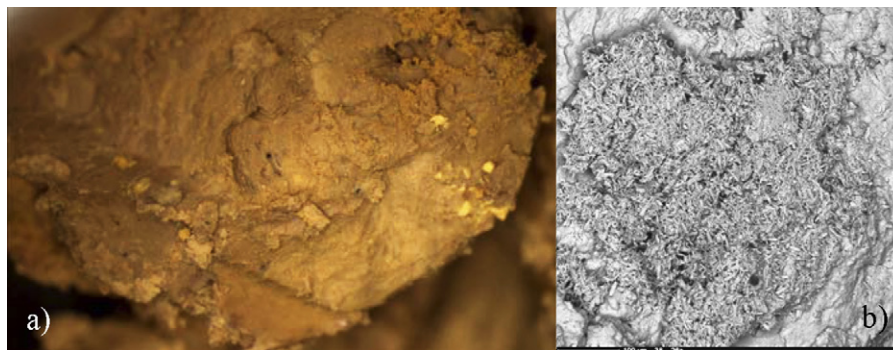


Fig. 3. (a) Optical microscopy picture of the material, and (b) SEM micrograph showing the crystalline and amorphous part around it in the sample.

as ^{137}Cs and ^{154}Eu , as well as ^{241}Am . This indicates that the material originates from a country, having facilities for irradiation of nuclear material and for handling and possibly also for processing of spent fuel.

The natural uranium did not show traces of ^{236}U nor of any fission products, i.e. the material has neither been irradiated in a reactor nor has it been subject to an enrichment process. A number of additional parameters have been adapted from other fields of science, e.g. from geology and can be used to either exclude certain origins or ideally to positively identify the geographical origin (so-called geolocation) of the uranium.

Uranium is typically recovered from uranium ore deposits where the concentrations vary from few per mille to several per cents in the very rich ores. Also phosphate ores contain U, though in relatively low concentrations (few hundred ppm). The large scale processing of phosphate ores for fertilizer production, allows for the recovery of uranium as a by-product. Irrespective of the origin of the uranium, all the major and minor accompanying elements are separated from U when it is processed, but traces of them will follow through the whole process from ore via yellow cake to the final product of U-oxide. Each process step reduces the impurity content and, on the other hand, may introduce new impurities, e.g. from processing tools, from reagents or from corrosion or abrasion of container walls. It has, however, been observed that the pattern of certain impurities remains unchanged throughout the process [13,21]. These parameters are characteristic for the starting material.

The isotopic composition of minor constituents may also provide useful information. The isotopic composition of lead is a parameter that has been noticed to vary as a function of the U mine to another or more generally from one source to another [14–20]. Three out of four of the stable Pb isotopes, namely ^{206}Pb , ^{207}Pb and ^{208}Pb , are the end products of the radioactive decay series of ^{238}U , ^{235}U and ^{232}Th , respectively. Therefore, depending on the age of the ore body and on the initial U/Th ratio in the mine, variations in the isotopic composition of Pb are found. In the present sample, the lead showed an isotopic composition close to natural lead. The absence of radiogenic lead points at a mineral of rather low uranium content, as for instance in phosphate ores.

The third possible signature for geolocation is the isotopic ratio of $^{18}\text{O}/^{16}\text{O}$. This signature is, however, useable only for U-

oxide samples. It is well documented that the $^{18}\text{O}/^{16}\text{O}$ ratio in surface waters varies around the world due to the natural isotopic fractionation [22,23]. There are several parameters, which cause variations of around 5% in the oxygen ratio, such as temperature, latitude and distance to the sea. As water is used in the wet processing of uranium, the final U-oxide product, e.g. UO_2 or U_3O_8 carries the signature of the $^{18}\text{O}/^{16}\text{O}$ ratio of the water [24,25]. This method was, however, not applied to the sample of yellow cake.

2.4. Application of classical forensic methods

Another major step was the connection between nuclear forensics (which focuses on the information inherent to the material) and classical forensics (which focuses on the information adherent to the material). Classical forensics basically serves for prosecution purposes and aims at identifying individuals and at establishing relations between locations, events and individuals. It is based on the “Locard principle”, which says whenever two objects meet, there is an exchange of material from each to the other (Edmond Locard, French scientist). The most common traces of material exchanged on a crime scene and investigated in forensic laboratories are fingerprints, DNA, fibres, hair, tool marks, glass, vegetation, soil or paint. Classical forensic investigations on radioactively contaminated items pose a serious problem to forensic laboratories. Dedicated installations, licensed for handling of radioactive material and providing the analyst sufficient protection against the radiation and the risk of incorporation are required. Such installations, e.g. for taking fingerprints from contaminated pieces of evidence have been developed and implemented in few nuclear laboratories. An example is shown in Fig. 4, where contaminated evidence can be visually inspected, photographed and fingerprints can be developed using the well established cyanacrylate (superglue) method. The glove-box and the associated protocols have been developed jointly by the Institute for Transuranium Elements and by the German federal criminal police.

The protocols for taking DNA samples from contaminated items, for isolation and extraction of the DNA are being developed. Also the radiation stability of the “genetic fingerprint” is being studied by various groups. Preliminary results show



Fig. 4. Glove-box for handling of nuclear material, equipped with a fuming chamber for visualising fingerprints using the cyanoacrylate method.

that the DNA can take doses of up to 5 kGy before the genetic fingerprint is wiped out [34].

3. Data interpretation and attribution

The main challenges in nuclear forensics are with the identification of characteristic parameters and with the availability of reference information. Based on reliable measurements of well-chosen parameters, clues on the origin of the material can be obtained. Information obtained by nuclear forensic analyses from an unknown nuclear material can basically be divided into two groups: endogenic and exogenic information. Endogenic information is to be understood as being self-explaining, and only some model calculations might be required to help data interpretation. Above in Section 2.1, presented parameters like isotopic compositions of U and Pu are good examples about endogenic information. They tell immediately about the material, in this case about its intended use, and additionally for Pu, the reactor type where the material was produced. Other endogenic parameters are, e.g. the U-content and the age of the material [26–30].

In contrast to that, exogenic information needs to be compared with data from known samples. The availability of “reference information” or comparison samples is essential for the interpretation of data like chemical impurities or the isotopic composition of minor constituents. This type of information includes all the geolocation and production parameters, i.e. impurities, Pb isotopic composition, $^{18}\text{O}/^{16}\text{O}$ ratio and microstructure. Known data can be either compiled and systematized in form of a (relational) database, where data e.g. from fuel manufacturers is collected, or it can be collection of analytical results of known samples [31–33]. An example of correlation using data obtained from the database of the ITU can be seen in Fig. 5. In this correlation Al and N were used to distinguish products from three different MOX fuels. One can notice that correctly chosen combination of impurities can be helpful in the origin determination or in the exclusion of certain fabrication plants or batches. The drawback of the use of impurities is

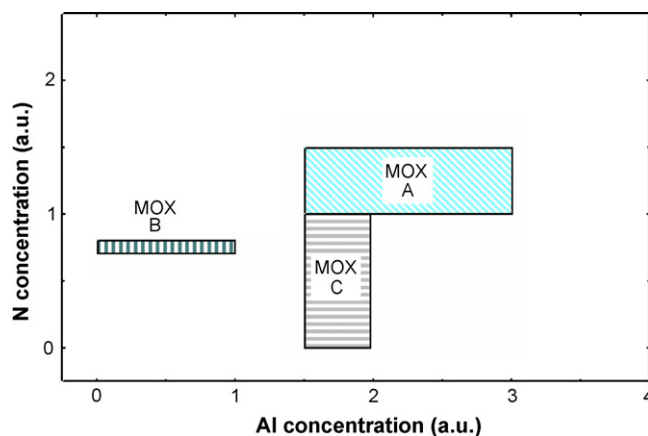


Fig. 5. Example of an impurity correlation for three different MOX fuels.

that one has to be very careful with cross-contamination (e.g. from environment). In addition the level of impurities within a production plant may vary with time, thus requiring continuous input from the production plants and as well as updating of the database.

A major challenge in data interpretation is the accessibility of reference data. Measured data on reactor grade nuclear material, which is essentially produced for power reactors, are available with the fuel manufacturers. ITU has established a nuclear materials database in collaboration with the A.A. Bochvar Institute in Moscow. This database contains information on nuclear fuels as collected from the open literature, as well as data provided by Russian and some European manufacturers. It has to be noted that certain data (e.g. chemical impurities) might be commercially sensitive and therefore their accessibility might be limited. Detailed information on weapons grade material (highly enriched uranium and low burn-up plutonium) is obviously subject to confidentiality for national security reasons and data are not shared.

Source attribution (i.e. the determination of the origin of the material) is generally done by comparing measurement results to data contained in a database applying the “exclusion principle”. The results of the first measurements on the unknown material (e.g. pellet dimensions and isotopic composition) are used for a query. All non-matching records (database entries from known materials) are rejected, the matching records are compared to each other in order to identify parameters to be analysed next. Thus, the database query also serves for analytical guidance, streamlining the laboratory work and rendering the measurement effort more efficient. Based on the subsequent measurement data, another query is performed in the database and further records are rejected. This process then results in records where all search parameters match within the stated tolerances. Ideally, the search results point at a single place of production.

4. Outlook

Nuclear forensic science is a fairly young discipline and only a small number of laboratories are active practitioners. However, the number of incidents of illicit trafficking reported to the

IAEA is reason for concern. Furthermore, the threat of nuclear terrorism calls for preparedness and for effective tools providing hints on the origin of the material and thus on the perpetrator.

The determination of characteristic parameters is subject to ongoing research and development work in a number of nuclear measurement laboratories. In particular, geolocation methods are to be refined and methodologies existing in other fields of science (geology, cosmology, etc.) should be transferred to the nuclear forensic area.

The availability of comparison data is of key importance for data interpretation. The International Atomic Energy Agency is collecting information on existing databases that could be useful in this respect. Data on nuclear fuels, on uranium ores, on weapons material, on commercial reactors, on research reactors and other relevant information have been collected by various institutions for different purposes. Although the data itself might not be shared, it would be most useful to establish a “database of databases” in order to possibly pose queries to those databases (offered by the respective owner on a voluntary basis).

International collaboration on nuclear forensics has been established already in 1996 with the creation of the International Technical Working Group on Nuclear Smuggling (ITWG) under the auspices of the Non-Proliferation Experts Group of the G-8. The ITWG provides a forum for nuclear forensic laboratories to exchange experience and to jointly advance this area of science by discussing analytical methods, evaluation and interpretation techniques and by defining the requirements for appropriate reference materials. In round robin exercises the laboratories’ capabilities are challenged and tested. Also the IAEA has addressed the issue of nuclear forensics and of providing nuclear forensic support to its member states and issued a technical guidance [35]. A recently launched “Co-ordinated Research Program” (CRP) of the IAEA aims at identifying gaps in the nuclear forensics knowledge and at addressing those gaps through dedicated research activities.

The co-ordinated application of classical forensics and of nuclear forensics to items under investigation needs to be fully established and appropriate protocols need to be developed. These protocols should cover the management of contaminated crime scenes, sample taking in a contaminated environment, preservation of both nuclear and classical forensic evidence, chain of custody, handling of evidence in a nuclear laboratory and writing expert witness reports.

Obviously, these activities need to be encompassed by appropriate training programs in order to increase the nuclear forensic awareness.

5. Conclusions

A new discipline in science was born in the early 1990s after the first cases of nuclear smuggling appeared: nuclear forensics. Created from sheer necessity, it rapidly developed to a powerful tool for identifying the possible origin of nuclear material and its intended use. The analytical approach and the interpretation of measurement data were systematized and today a methodology is in place which enables credible results. International cooperation has positively advanced this development and nuclear

forensic capabilities are available in several countries. Parameters like isotopic composition, chemical impurities, age of the material, macroscopic parameters and microstructure provide clues on the origin and on the intended use of the material. A wide variety of analytical techniques, specifically adapted for measuring nuclear material is used for investigating nuclear material intercepted from illicit incidents. Still, there is no silver bullet, no single parameter that would point at the source of the material. Source attribution requires the determination of a characteristic pattern of parameters and the availability of reference data for comparison purposes.

The main challenges in the area of nuclear forensics are

- The identification of additional parameters that are characteristic for the origin of a material, for the starting material used for its production or for the type of production process applied.
- The accessibility of databases for comparing data obtained on seized material to data from material of known origin and history.

Today, nuclear forensics has reached a high degree of maturity and it is highly relevant in the areas of non-proliferation and of nuclear security. Continued development activities and strengthened international cooperation will be of key importance to perfection the discipline of nuclear forensics.

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